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**Title of the Invention: Polyester-based Resin Composition For
Calendering and Film or Sheet Made Using This Resin Composition**

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(54) [Title of the Invention]

**Polyester-based Resin Composition For
Calendering and Film or Sheet Made Using
This Resin Composition**

(57) [Summary]

[Object] To provide a polyester-based resin composition that has good release from metal rolls, good take-off by cooling rolls, and good rotating properties of the bank; does not entrain air into the bank; reduces the load on the processing machines during calendering; and allows a film or sheet that has little shrinkage and possesses excellent transparency, rigidity, gas barrier properties, and wettability to be obtained by calendering.

[Means of Achievement] An organically modified layered silicate, a fatty acid having 18 or more carbon atoms, and a fatty acid metal salt or organic phosphoric acid ester are admixed into a thermoplastic polyester resin of controlled crystallinity.

[Selected Drawing] None

[Claims]

[Claim 1] A polyester-based resin composition for calendering, characterized in that 100 parts by weight of an amorphous or low-crystallinity polyester resin (A) that has no crystal melting peak or a crystal melting peak at 200°C or lower and a crystal heat of fusion of 50 J/g or less when measured by DSC (differential scanning calorimeter) are mixed with 0.5-20 parts by weight of organically modified layered silicate (B), 0.1-10 parts by weight of a fatty acid with 18 or more carbon atoms (C), and 0.01-5 parts by weight of fatty acid metal salt (D).

[Claim 2] A polyester-based resin composition for calendering, characterized in that 100 parts by weight of the amorphous or low-crystallinity polyester resin (A) according to Claim 1 are mixed with 0.5-20 parts by weight of organically modified layered silicate (B), 0.1-10 parts by weight of a fatty acid with 18 or more carbon atoms (C), and 0.01-5 parts by weight of organic phosphoric acid ester (E).

[Claim 3] The polyester-based resin composition for calendering according to Claim 1 or 2 characterized in that the polyester-based resin (A) is a polyester-based resin composed of repeating units of a dicarboxylic acid component 80 mol% or more of which is one type or a mixture of two or more types selected from among terephthalic acid, naphthalenedicarboxylic acid, 1,4-cyclohexanedicarboxylic acid, and isophthalic acid; and a glycol component 80 mol% or more of which is one type or a mixture of two or more types selected from among ethylene glycol, neopentyl glycol, diethylene glycol, and 1,4-cyclohexanedimethanol.

[Claim 4] The polyester-based resin composition for calendering according to any of Claims 1 to 3, characterized in that the organically modified layered silicate (B) is a silicate with intercalated organic onium ions.

[Claim 5] The polyester-based resin composition for calendering according to any of Claims 1 to 4, characterized in that the ratio (c'/b') of the parts by weight (b') of silicate in the organically modified layered silicate added per 100 parts by weight of the polyester-based resin (A) according to Claim 1, and the parts by weight (c') of the fatty acid with 18 or more carbon atoms added per 100 parts by weight of the same polyester-based resin (A) is 0.1 or higher.

[Claim 6] A film or sheet formed by calendering using the thermoplastic polyester resin composition for calendering according to any of Claims 1 to 5.

[Claim 7] A film or sheet with a thickness of 0.2 mm or less formed by calendering using the thermoplastic polyester resin composition according to any of Claims 1 to 5, wherein said film or sheet is characterized in that the shrinkage index of a molded article heat-treated for 60 minutes at a temperature 10°C higher than the glass transition temperature of the molded article is less than 10%; the shrinkage index of a molded article heat-treated for 60 minutes at a temperature 20°C higher than the glass transition temperature of the molded article is less than 15%; and the shrinkage index of a molded article heat-treated for 60 minutes at a temperature 40°C higher than the glass transition temperature of the molded article is less than 20%.

[Detailed Description of the Invention]

[0001]

[Technological Field of the Invention]

The present invention relates in particular to a polyester-based resin composition molded by calendering, and to a film or sheet obtained by calendering this resin composition.

[0002]

[Prior Art]

Thermoplastic polyester resins such as polyethylene terephthalate (PET) and polybutylene terephthalate (PBT) are widely used as packaging materials for foods and drug products, materials for container lids and blister packaging, and as laminated films and sheets for construction, consumer electronics, and automobiles because of their excellent mechanical properties, heat resistance, and gas barrier properties.

[0003]

Extrusion molding is commonly used when films and sheets are molded using thermoplastic polyester resins. In extrusion molding, molten resin is discharged by utilizing the extrusion force of a screw after the die has been adjusted so that the sheet will have a specific thickness, and the sheet is taken off while the molten resin is rapidly cooled to a temperature lower than its softening temperature. The melt is therefore easy to take off by the cooling rolls, but parts of different thicknesses tend to develop even when the die is adjusted, and the thickness accuracy of the sheet is inferior. This tends to cause problems in subsequent processing such as

printing, lamination, and coating, and holes open in the sheet in secondary molding such as vacuum and compression molding. Furthermore, the molding speed is low when a sheet is molded by extrusion molding, and it is difficult to regard the productivity as satisfactory.

[0004]

For such reasons, calendering is often used instead of extrusion molding to produce sheets and films. Calendering is a molding method that produces a sheet or film of the desired thickness by rolling the molten resin using a plurality of heated metal rolls (calender rolls) and then taking off the sheet or film onto cooled rolls. There are none of the problems that develop near the die in extrusion molding, the thickness accuracy of the sheet or film molded is good, and a sheet or film of excellent quality can be produced. The molding speed is also high and the productivity is excellent. This method is therefore suited to the mass production of articles of the same scale.

[0005]

However, films and sheets cannot be produced from thermoplastic polyester resins such as PET and PBT by calendering. This is because, unlike in processing methods in which a film or sheet is molded by utilizing cooling rolls to take off the molten resin discharged from the die by the extrusion force of a screw in the way it is done in extrusion molding, calendering is a processing method in which a film or sheet is molded by utilizing cooling rolls to take off the molten resin rolled by a plurality of heated calender rolls while the tension of the molten resin is used and the resin is released from the calender rolls. High take-off resistance is therefore required of the molten resin.

In other words, thermoplastic polyester resins such as PET and PBT generally must be molded at a temperature of 250°C or higher. Since this type of thermoplastic polyester resin has very low melt viscosity and melt tension at a temperature of 250°C or higher, it does not have high take-off resistance, and calendering therefore cannot be applied to it.

[0006]

To address this problem, attempts have been made to increase the take-off resistance to make the resin suitable for calendering by controlling the crystallinity through the production of a random copolymer according to a method in which part of the terephthalic acid or ethylene glycol monomer is substituted by another component to allow molding to be performed at a temperature of about 200°C in the case of PET-based resin, which is one of thermoplastic

polyester resins. PET-based resins, however, stick severely to the calender rolls because of their high polarity, and the resin itself cannot be used in calendering even if the take-off resistance has been increased.

[0007]

Mixing of various additives has therefore been proposed in JP Kokai 11-343353, 2000-186191, 2000-302951, 2000-327891, and 2001-64496 as a method of making it easier to release the resin from the calender rolls.

[0008]

Additives that have low affinity for the resin are generally deemed to be effective for attaining easy release from the metal rolls (calender rolls). Introducing such additives, however, lowers the transparency of the molded sheet or film and sometimes causes the materials to bleed out onto the metal rolls. It was consequently extremely difficult to make it easier for the resin to be released from the metal rolls without harming the transparency and causing the materials to bleed out onto the metal rolls even when a PET-based polyester resin was calendered by the disclosed techniques of introducing additives into polyester-based resins modified so as to be suitable for calendering by controlling resin crystallinity.

[0009]

Moreover, even if a state in which calendering is possible is attained by introducing additives to facilitate release, the high melt viscosity of the resin molded at a temperature that permits calendering and yields the desired take-off resistance causes the driving power of the calendering equipment to be increased. As a result, the cost of electrical power needed for production increases and production problems arise in molding equipment with low drive motor power.

[0010]

On the other hand, thin articles are generally produced in an efficient manner when calendering is performed by melt drawing in which the take-off speed of the cooling rolls is raised above the rotational speed of the calender rolls. The reasons are that if the gap between the calender rolls is too narrow, there is a risk that the calender rolls will come into contact with each other, adjusting the gap between the calender rolls alone produces limited results in terms of the production of thin films and sheets because the resin melt swells after being rolled, and the molten resin tends to evolve heat and adhere to the calender rolls. Another reason is that

performing melt drawing by setting the take-off speed of the cooling rolls above the rotational speed of the calender rolls increases the production speed.

[0011]

The problem with films and sheets produced by calendering via a melt drawing step is that significant shrinkage occurs in the direction of melt drawing in steps where the film or sheet is reheated, such as during drying or application of printing ink or an adhesive agent. The product also shrinks and the same problems are encountered with films and sheets that are used in heat insulation and in displays, and with films that are used to prevent glass from scattering and attached in locations with relatively high temperatures, such as in windows. Such shrinkage is especially evident in the case of thermoplastic polyester resins.

[0012]

Methods of adding glass fibers and fillers such as calcium carbonate and talc were used in the past to improve the dimensional stability, mechanical properties, heat resistance, and gas barrier properties of thermoplastic polyester resins such as PET and PBT. These conventional methods have the drawbacks of increasing the specific gravity of the resulting film or sheet, compromising the surface appearance, and making it impossible to obtain a transparent product.

[0013]

A technique that relates to compositions and composite materials in which smectite-based clay minerals such as montmorillonite, and organically modified layered silicate in which interlayer ions are ion-exchanged by organic onium ions, such as fluorinated mica, are finely dispersed in a thermoplastic polyester resin is known as a method that can improve upon such drawbacks and enhance the dimensional stability, mechanical properties, heat resistance, and gas barrier properties of films and sheets made from thermoplastic polyester resins.

[0014]

For example, JP Kokai 3-62846 discloses a polyester composite material that contains thermoplastic aromatic polyester, a layered clay mineral bonded with organic onium ions, and a compatibilizing agent, and has a structure which is crosslinked by the molecular chains of the thermoplastic aromatic polyester and in which the layered clay mineral bonded with organic onium ions is uniformly dispersed in the thermoplastic aromatic polyester by the affinity action of the compatibilizing agent, and also discloses a process for the production of this material.

JP Kokai 7-166036 also discloses an aromatic polyester resin that contains 0.5-10 parts by weight (in terms of inorganic ash) of an intercalation compound that has layered silicate with a cation exchange capacity of 30 mEq/100 g or more as the host, and a quaternary ammonium ion as the guest. JP Kokai 2000-53847 discloses a thermoplastic polyester resin composition in which the content of carboxyl end groups is 10-80 Eq/t and which is obtained by melt kneading thermoplastic polyester and a layered silicate in which intercalated exchangeable cations have been exchanged by organic onium ions. JP Kokai 2000-212424 discloses a polyester resin composition obtained by mixing 0.1-40 parts by weight of a layered silicate (in which the intercalated exchangeable cations have been exchanged by organic onium ions) and 0.001-5 parts by weight of montanic acid and/or a low-molecular-weight polyolefin.

[0015]

However, even with the techniques disclosed in relation to composite materials and compositions obtained using thermoplastic polyester resins, a thermoplastic polyester resin that has excellent dimensional stability, mechanical properties, heat resistance, and gas barrier properties is still difficult to produce by calendering, which requires high take-off resistance. Furthermore, no mention at all is made in the aforementioned prior art literature regarding methods that allow shrinkage, which is a particular problem, to be reduced in the manufacture of films and sheets from thermoplastic polyester resins by calendering.

[0016]

[Disclosure of Prior Art References]

[Prior Art 1]

JP Kokai Hei 3-62846

[Prior Art 2]

JP Kokai Hei 7-166036

[Prior Art 3]

JP Kokai 2000-53847

[Prior Art 4]

JP Kokai 2000-212424

[0017]

[Problems to Be Solved by the Invention]

The present invention, which was perfected in view of this situation, is aimed at providing a thermoplastic polyester resin composition that can be readily molded by calendering, has excellent transparency, rigidity, gas barrier properties, and wettability as a molded resin, and yields films and sheets characterized by reduced shrinkage due to molding; and to provide films and sheets obtained by calendering this resin composition.

[0018]

[Means Used to Solve the Above-Mentioned Problems]

As a result of diligent studies conducted in order to resolve the above problems, the inventors perfected the present invention upon discovering that films and sheets with excellent calendering moldability, reduced shrinkage due to molding, and excellent transparency, rigidity, gas barrier properties, and wettability can be produced by using a resin composition that combines specific numbers of parts by weight of specific additives with a specific thermoplastic polyester resin.

[0019]

Specifically, the polyester-based resin composition for calendering according to Claim 1 of the present invention is characterized in that 100 parts by weight of an amorphous or low-crystallinity polyester resin (A) that has no crystal melting peak or a crystal melting peak at 200°C or lower and a crystal heat of fusion of 50 J/g or less when measured by DSC (differential scanning calorimeter) are mixed with 0.5-20 parts by weight of organically modified layered silicate (B), 0.1-10 parts by weight of a fatty acid with 18 or more carbon atoms (C), and 0.01-5 parts by weight of fatty acid metal salt (D).

[0020]

The term "DSC" used in the present invention is an abbreviation for a differential scanning calorimeter or measurement method. It means an apparatus or measurement method used to measure the melting point and heat of fusion, and is the one used in JIS K-7121 and K-7122. The crystal melting peak and crystal heat of fusion were measured in accordance with JIS K-7121 and K-7122 as well.

[0021]

The polyester-based resin composition for calendering according to Claim 2 is characterized in that 100 parts by weight of the amorphous or low-crystallinity polyester resin (A) according to Claim 1 are mixed with 0.5-20 parts by weight of organically modified layered silicate (B), 0.1-10 parts by weight of a fatty acid with 18 or more carbon atoms (C), and 0.01-5 parts by weight of organic phosphoric acid ester (E).

It is most preferable that the amorphous or low-crystallinity polyester resin (A) be a polyester-based resin that comprises repeating units of a dicarboxylic acid component 80 mol% or more of which is one type or a mixture of two or more types selected from among terephthalic acid, naphthalenedicarboxylic acid, 1,4-cyclohexanedicarboxylic acid, and isophthalic acid; and a glycol component 80 mol% or more of which is one type or a mixture of two or more types selected from among ethylene glycol, neopentyl glycol, diethylene glycol, and 1,4-cyclohexanedimethanol (Claim 3).

It is also preferable that the organically modified layered silicate (B) be a resin with intercalated organic onium ions (Claim 4).

The ratio (c'/b') of the parts by weight (b') of silicate in the organically modified layered silicate added per 100 parts by weight of the polyester-based resin (A) according to Claim 1 and the parts by weight (c') of the fatty acid with 18 or more carbon atoms added per 100 parts by weight of the same polyester-based resin (A) is preferably 0.1 or higher (Claim 5).

The film or sheet (Claim 6) molded by calendering using the thermoplastic polyester resin composition for calendering according to any of Claims 1-5 is characterized in that a film or sheet heat-treated for 60 minutes at a temperature 10°C higher than the glass transition temperature of the molded article has a thickness of 0.2 mm or less and the shrinkage index of the molded article is less than 10%, that the shrinkage index of a molded article heat-treated for 60 minutes at a temperature 20°C higher than the glass transition temperature of the molded article is less than 15%, and that the shrinkage index of a molded article heat-treated for 60 minutes at a temperature 40°C higher than the glass transition temperature of the molded article is less than 20% (Claim 7).

[0022]

[Embodiments of the Invention]

Embodiments of the present invention are explained below.

An amorphous or low-crystallinity polyester resin that has no crystal melting peak or a crystal melting peak of 200°C or lower and a crystal heat of fusion of 50 J/g or lower when measured by DSC is used as the polyester-based resin (A) in the polyester-based resin composition for calendering of the present invention.

[0023]

A polyester-based resin whose crystal melting peak is greater than 200°C melts only when the temperature exceeds 200°C. It therefore becomes difficult to mold a film or sheet by calendering because the melt viscosity and melt tension of the polyester-based resin become extremely low when the resin is molded in this temperature region.

The crystal heat of fusion of the polyester-based resin must be 50 J/g or lower to minimize the development of problems in which the unmelted resin is left behind as a gel in the form of a film or sheet, particularly when calendering is used. A crystal heat of fusion of 30 J/g or lower is preferred, and 10 J/g or lower is more preferred. It is most preferable to use an amorphous polyester resin that has essentially no crystal melting peak because such a resin can be melted at a low temperature.

[0024]

In addition to a polyester-based resin of a unitary composition that fulfills the above requirements, mixtures of multiple polyester-based resins and mixtures of 50% by weight or more of polyester-based resin and resins other than polyester-based resins, such as polyolefin-based resins, polyamide-based resins, and vinyl chloride-based resins, can also be used as the polyester-based resin (A). However, a polyester-based resin having a unitary composition is suitable as the polyester-based resin (A) used in the present invention because of the requirements of excellent calendering moldability and transparency of the resulting film or sheet.

[0025]

The polyester-based resin (A) used in the present invention can be obtained by copolymerizing one or more types of carboxylic acid component and/or one or more types of glycol component.

Examples of the carboxylic acid component include dicarboxylic acids such as terephthalic acid, naphthalenedicarboxylic acid, 1,4-cyclohexanedicarboxylic acid, isophthalic acid, succinic acid, glutaric acid, adipic acid, sebacic acid, suberic acid, azelaic acid, dodecanedioic acid, fumaric acid, maleic acid, itaconic acid, o-toluic acid, 2,6-naphthalene

dicarboxylic acid, 2,6-naphthalene didimethylene [sic] carboxylic acid, and p-phenylene dicarboxylic acid; tricarboxylic acids such as trimellitic acid; and tetracarboxylic acids such as pyromellitic acid.

[0026]

Examples of the glycol component include dihydric alcohols such as ethylene glycol, neopentyl glycol, diethylene glycol, 1,4-cyclohexanedimethanol, propylene glycol, 1,3-propanediol, 2,4-dimethyl-2-ethylhexane-1,3-diol, 2,2-dimethyl-1,3-propanediol, 2-ethyl-2-butyl-1,3--propanediol, 2-ethyl-2-isobutyl-1,3-propanediol, 1,3-butanediol, 1,4-butanediol, neopentyl glycol, 1,5-pentanediol, 1,6-hexanediol, 1,8-octanediol, 2,2,4-trimethyl-1,6-hexanediol, thiodiethanol-1,2-cyclohexanedimethanol, 1,3-cyclohexanedimethanol, and 2,2,4,4-tetramethyl-1,3-cyclobutanediol; trihydric alcohols such as trimethylolpropane; and tetrahydric alcohols such as pentaerythritol.

[0027]

In the polyester-based resin used in the present invention, 80 mol% or more of the carboxylic acid component is preferably one type or a mixture of two or more types selected from among terephthalic acid, naphthalenedicarboxylic acid, 1,4-cyclohexanedicarboxylic acid, and isophthalic acid, and 80 mol% or more of the glycol component is preferably one type or a mixture of two or more types selected from among ethylene glycol, neopentyl glycol, diethylene glycol, and 1,4-cyclohexanedimethanol.

Among these, it is preferable to use a polyester-based resin that has a dicarboxylic acid component comprising 90 mol% or more terephthalic acid, and a glycol component comprising 10-95 mol% 1,4-cyclohexanedimethanol and 90-5 mol% ethylene glycol, and components that are readily available as raw materials can be used. It is most preferable to use an amorphous polyester resin made from these components.

This is because the polyester-based resin can form a melt at a low temperature if it is amorphous. This is advantageous for calendering that requires especially high melt tension and also gives good surface smoothness and transparency to the sheet or film obtained by calendering.

[0028]

The amorphousness of the polyester-based resin varies depending on the composition ratio of the monomer components, but 100 mol% terephthalic acid, 30-90 mol% ethylene glycol, and 10-70 mol% 1,4-cyclohexanedimethanol are preferred in order to attain amorphousness and

to simultaneously provide good moldability in calendering. Especially preferred is a composition comprising 100 mol% terephthalic acid, 60-75 mol% ethylene glycol, and 40-25 mol% 1,4-cyclohexanedimethanol.

[0029]

The polyester-based resin used in the present invention is preferably obtained by reacting a dicarboxylic acid component that contains terephthalic acid, and a glycol component that contains 1,4-cyclohexanedimethanol and ethylene glycol, and then polycondensing the components in the presence of a specific catalyst. Specifically, a dicarboxylic acid component that contains at least 90 mol% terephthalic acid, and a glycol component that contains 10-95 mol% 1,4-cyclohexanedimethanol and 90-5 mol% ethylene glycol are reacted at a temperature sufficient to induce esterification or transesterification. The polyester-based resin is then obtained as a colorless, transparent copolyester by polycondensing the reaction product for less than 2 hours at an absolute pressure lower than 1.333 kPa in the presence of an inhibitor and a catalyst composed of 0-75 ppm Mn, 50-150 ppm Zn, 5-20 ppm Ti, 5-200 ppm Ge, and 10-80 ppm P, based on the weight of the copolyester.

[0030]

When the resin is molded into a film or sheet, crosslinking may be performed within the range that does not negatively affect the transparency in order to raise the melt tension and to simultaneously improve the calendering moldability of the polyester-based resin obtained in this way. The crosslinking is performed by reaction with a crosslinkable compound during the polycondensation of the polyester-based resin or after the resin has been polycondensed.

[0031]

Some resins have already been marketed as polyester-based resins that can be used in the present invention. For example, amorphous polyester resins composed of 100 mol% terephthalic acid, 60-75 mol% ethylene glycol, and 40-25 mol% 1,4-cyclohexanedimethanol are marketed by Eastman Chemical Products as Kodar PETG Copolyester (trade name) and Tsunami Copolyester (trade name). Such polyester-based resins can be used appropriately in the present invention.

[0032]

The molecular weight of the amorphous or low-crystallinity polyester resin (A) used in the present invention is not particularly restricted, but the melt tension of the molten resin is low and molding is difficult because the temperature range at which molding can be performed by

calendering is narrow when the number-average molecular weight is less than 10,000. The surface smoothness of the sheet or film obtained can be inferior when the number-average molecular weight exceeds 200,000. Therefore, the number-average molecular weight is preferably in the range of 10,000-200,000 in consideration of the moldability by calendering and the improved surface smoothness of the resulting sheet or film.

[0033]

The organically modified layered silicate (B) used in the present invention is a component admixed in order to improve performance attributes such as the rigidity, gas barrier properties, and wettability of the film or sheet obtained by calendering, and in order to reduce the shrinkage of the molded film or sheet together with improving the moldability in calendering. Compounds in which the exchangeable cations present between the layers of the layered silicic acid are substituted by organic onium ions can be used appropriately.

[0034]

The layered silicate with intercalated exchangeable cations has a laminated structure with a thickness of 0.5-2 nm and a width of 0.02-2 μm . The exchangeable cations of alkali metals such as Li^+ , Na^+ , and K^+ and alkaline earth metals such as Ca^{2+} and Mg^{2+} are present in a state hydrated by water molecules between the layers. The cation exchange capacity is preferably 30-300 mEq/100 g, more preferably 50-150 mEq/100 g.

The molded film or sheet becomes opaque and the effects of the present invention are not obtained because the layers of the layered silicate aggregate in the resin without separating due to inadequate onium ion exchange when the cation exchange capacity is less than 30 mEq/100 g. The molded film or sheet becomes opaque and the effects of the present invention are not obtained due to similar aggregation in the resin because of the strong bonding force between the layers of the layered silicate when the cation exchange capacity exceeds 300 mEq/100 g.

[0035]

Specific examples of the layered silicate with intercalated exchangeable cations include smectite-based clay minerals such as montmorillonite, saponite, hectorite, fluorohectorite, bidelite, and stevensite, and synthetic micas such as Li-fluoroteniolite, Na-fluoroteniolite, Na-tetrasilicic fluoromica, and Li-tetrasilicic fluoromica, as well as vermiculite, fluorovermiculite, halosite, kanemite, keniyte, zirconium phosphate, and titanium phosphate. Both natural and synthetic products are acceptable.

Smectite-based clay minerals such as montmorillonite, saponite, hectorite, fluorohectorite, bidelite, and stevensite, and synthetic micas such as Li-fluoroteniolite, Na-fluoroteniolite, Na-tetrasilicic fluoromica, and Li-tetrasilicic fluoromica are preferred insofar as they are materials readily available in the industry.

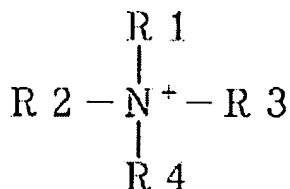
[0036]

Examples of the organic onium ions exchanged with the exchangeable cations of the layered silicate include ammonium ions, phosphonium ions, and sulfonium ions. Ammonium ions and phosphonium ions among them are preferred insofar as they are materials readily available in the industry. Ammonium ions are especially preferred.

[0037]

Specific examples of the ammonium ion include compounds shown by the following general formula

[Chemical Formula 1]



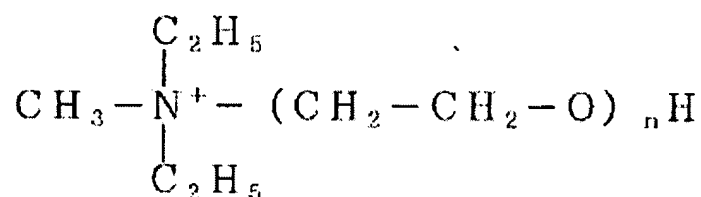
(where R¹-R⁴ are selected from among hydrogen, alkyl groups having 1-30 carbon atoms, benzyl groups, hydroxyalkyl groups having 1-30 carbon atoms, hydroxybenzyl groups, -(CH₂-CH₂-O)_n-H groups, and -(CH₂-CH(CH₃)-O)_m-H groups (where n and m are integers of 1-30)); compounds derived from aniline, p-phenylene diamine, α-naphthyl amine, p-aminodimethyl aniline, benzidine, pyridine, piperidine, 4-aminododecanoic acid, 6-aminocaproic acid, 12-amino-dodecanoic acid, and 18-aminooctadecanoic acid; and benzethonium ion.

[0038]

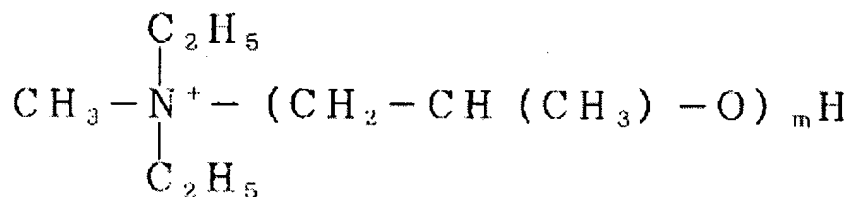
Specific examples of ammonium ion compounds shown by the above general formula include stearyl ammonium ion, dodecyl ammonium ion, benzyl ammonium ion, hexyl ammonium ion, octyl ammonium ion, 2-ethylhexyl ammonium ion, lauryl ammonium ion, methyl stearyl ammonium ion, distearyl ammonium ion, methyl distearyl ammonium ion, trioctyl ammonium ion, dimethylstearyl benzyl ammonium ion, methyl distearyl benzyl ammonium ion, distearyl dibenzyl ammonium ion, benzalkonium ion, trimethylstearyl ammonium ion, dimethyl distearyl ammonium ion, dimethyl 2-ethylhexyl stearyl ammonium ion, methyl tristearyl

ammonium ion, di-tallow alkyl dimethyl ammonium ion, di-coconut oil alkyl dimethyl ammonium ion, dimethyl dihydroxyethyl ammonium ion, dimethyl benzyl hydrogenated tallow ammonium ion, dimethyl di-hydrogenated tallow ammonium ion, methyl tallow alkyl bis-2-hydroxyethyl ammonium ion, methyl stearyl bis-2-hydroxyethyl ammonium ion, dimethyl hydrogenated tallow benzyl ammonium ion, dimethyl 2-ethylhexyl hydrogenated tallow ammonium ion, methyl di-hydrogenated tallow ammonium ion, and compounds shown by the following formulas (where m and n are integers of 1-30)

[Chemical formula 2]



[Chemical formula 3]



The ammonium ions introduced into the layered silicate by ion exchange may be individual ions or a mixture of multiple ions.

[0039]

Among these ammonium ions, organic ammonium ions with substituents such as alkyl groups having 6-30 carbon atoms, benzyl groups, and hydroxyalkyl groups are preferred. Trimethyl stearyl ammonium ion, dimethyl distearyl ammonium ion, dimethyl stearyl benzyl ammonium ion, methyl distearyl benzyl ammonium ion, benzalkonium ion, dimethyl benzyl hydrogenated tallow ammonium ion, dimethyl di-hydrogenated tallow ammonium ion, methyl tallow alkyl bis-2-hydroxyethyl ammonium ion, and methyl stearyl bis-2-hydroxyethyl ammonium ion are especially preferred.

[0040]

The amount of organic onium ion in the organically modified layered silicate is determined by the cation exchange capacity of the layered silicate used and the type of organic onium ion, but is usually in the range of 15-80% by weight.

[0041]

The amount in which the organically modified layered silicate (B) is added is in the range of 0.5-20 parts by weight, preferably 1.0-15.0 parts by weight, more preferably 2.0-13.0 parts by weight, per 100 parts by weight of the amorphous or low-crystallinity polyester resin (A). The improvement of moldability in calendaring is lacking and there is a risk that performance attributes of the resulting film or sheet, such as strength, rigidity, gas barrier properties, and shrinkability, will not be satisfactory when the amount in which the organically modified layered silicate (B) is added is less than 0.5 part by weight. There is a risk that the film or sheet will become brittle when the amount in which the organically modified layered silicate (B) is added exceeds 20 parts by weight.

[0042]

The fatty acid with 18 or more carbon atoms (C) used in the present invention is a component admixed in order to improve the releasability of the resin from the calender rolls during calendaring and the dispersiveness of the organically modified layered silicate (B). Examples include stearic acid, behenic acid, tetracosanoic acid, hexacosanoic acid, heptacosanoic acid, montanic acid, triacontanoic acid, and dotriacontanoic acid.

These fatty acids are available as highly pure reagents, but some are often expensive and are not commonly used as additives for resin compositions. However, stearic acid and additives for resins, such as behenic acid and montanic acid, can be obtained inexpensively as fatty acids with 18 or more carbon atoms (C), and such fatty acids for resin additives can also be used appropriately in the present invention.

[0043]

Behenic acid and montanic acid, which are fatty acids of 20 or more carbon atoms and can be obtained inexpensively as additives for resins, are preferred among these fatty acids because they resist degradation and evaporation at the temperature at which the thermoplastic polyester resin is molded. Montanic acid, which is a fatty acid of 25 or more carbon atoms and

can be obtained inexpensively as an additive for resins, is more appropriate for use in the present invention.

[0044]

The amount in which the fatty acid with 18 or more carbon atoms (C) is added per 100 parts by weight of polyester-based resin (A) is in the range of 0.1-10.0 parts by weight, preferably 0.3-7.5 parts by weight, more preferably 0.5-5.0 parts by weight. The releasability of resins from the calender rolls becomes unsatisfactory, dispersion of the organically modified layered silicate (B) becomes inadequate, and a film or sheet with excellent transparency cannot be obtained when the amount in which the fatty acid with 18 or more carbon atoms (C) is added is less than 0.1 part by weight. Not only does the transparency of the sheet or film drop, but it may also become impossible to produce a film or sheet by calendering due to excessive smoothness when the amount in which the fatty acid with 18 or more carbon atoms (C) is added exceeds 10.0 parts by weight.

[0045]

The ratio (c'/b') of the number of parts by weight (b') of silicate in the organically modified layered silicate (B) added per 100 parts by weight of the amorphous or low-crystallinity polyester resin (A), and the number of parts by weight (c') of the fatty acid with 18 or more carbon atoms (C) added per 100 parts by weight of the same polyester-based resin (A) is preferably 0.1 or higher, more preferably 0.15 or higher; in the resin composition of the present invention. An even greater effect is obtained with the optimum value of 0.2 or higher.

[0046]

The term "parts by weight (b') of silicate in the organically modified layered silicate (B)" used herein refers to a numerical value determined by the formula below from the percent by weight of ash (ash(%)) in the organically modified layered silicate, as determined based on the weight of the ash remaining after the organically modified layered silicate (B) has been heated for 30 minutes in a 1000°C electric furnace to evaporate and combust the water content and the organic onium ions contained between the layers of the organically modified layered silicate, and from the parts by weight (b'') of the organically modified layered silicate (B) added per 100 parts by weight of the amorphous or low-crystallinity polyester resin (A).

$$b' = b'' \times \text{ash} (\%) - 100$$

The organically modified layered silicate disperses inadequately in the polyester-based resin, and it becomes difficult to attain the goals of the present invention when the ratio (c'/b') of the parts by weight (b') of silicate in the organically modified layered silicate added per 100 parts by weight of the polyester-based resin (A) and the parts by weight (c') of the fatty acid with 18 or more carbon atoms added per 100 parts by weight of the same polyester-based resin (A) is less than 0.1.

[0047]

The fatty acid metal salt (D) used in the present invention is a component added to prevent air from being entrained into the molten resin mass (usually termed "the bank") formed between the calender rolls during calendaring, and to prevent the air from forming longitudinal streaks on the surface of the sheet or film.

[0048]

The number of carbon atoms in the fatty acid moiety is not particularly restricted when a fatty acid metal salt (D) is admixed. Examples of fatty acid metal salts that can be used include lithium stearate, magnesium stearate, calcium stearate, potassium stearate, aluminum stearate, strontium stearate, barium stearate, cadmium stearate, zinc stearate, tin stearate, calcium laurate, barium laurate, cadmium laurate, zinc laurate, lithium myristate, zinc oleate, calcium behenate, zinc behenate, magnesium behenate, lithium behenate, calcium montanate, magnesium montanate, sodium montanate, cadmium 2-ethylhexoate, and zinc 2-ethylhexoate. These fatty acid metal salts are marketed as resin additives and are appropriate for use in terms of cost.

[0049]

The term "organic phosphoric acid ester (E) used in the present invention" refers to an organic phosphoric acid ester or a partially saponified organic phosphoric acid ester. These can be used individually or in mixtures of two or more types.

[0050]

The type of organic phosphoric ester (E) is not particularly restricted. Examples include monostearyl phosphate, distearyl phosphate, tristearyl phosphate, monolauryl phosphate, dilauryl phosphate, trilauryl phosphate, monononylphenyl polyoxyethylene phosphate, dinonylphenyl polyoxyethylene phosphate, trinonylphenyl polyoxyethylene phosphate, monolauryl polyoxyethylene phosphate, dilauryl polyoxyethylene phosphate, trilauryl polyoxyethylene phosphate, monodecyl polyoxyethylene phosphate, dodecyl polyoxyethylene phosphate, tridecyl polyoxy-

ethylene phosphate, monododecyl polyoxyethylene phosphate, didodecyl polyoxyethylene phosphate, tridodecyl polyoxyethylene phosphate, monooctylphenyl polyoxyethylene phosphate, dioctylphenyl polyoxyethylene phosphate, trioctylphenyl polyoxyethylene phosphate, monododecylphenyl polyoxyethylene phosphate, didodecylphenyl polyoxyethylene phosphate, tridodecylphenyl polyoxyethylene phosphate, monostearyl dilauryl phosphate, monolauryl distearyl phosphate, monostearyl dinonylphenyl polyoxyethylene phosphate, monononylphenyl polyoxyethylene distearyl phosphate, monolauryl dinonylphenyl polyoxyethylene phosphate, monononylphenyl polyoxyethylene dilauryl phosphate, monostearyl dilauryl polyoxyethylene phosphate, monolauryl polyoxyethylene distearyl phosphate, monolauryl dilauryl polyoxyethylene phosphate, and monolauryl polyoxyethylene dilauryl phosphate. These can be used individually or in mixtures of two or more types. These organic phosphoric acid esters are marketed as resin additives and are appropriate for use in terms of cost.

[0051]

The type of partially saponified organic phosphoric acid ester used is also not particularly limited. Examples include the aforementioned mono- and di-phosphoric acid esters saponified by the remaining acid radicals. Specific examples include calcium-, magnesium-, and zinc-saponified forms of the above organic phosphoric acid esters, e.g., calcium saponified monostearyl phosphate, magnesium saponified monostearyl phosphate, zinc saponified monostearyl phosphate, calcium saponified monolauryl phosphate, magnesium saponified monolauryl phosphate, zinc saponified monolauryl phosphate, calcium saponified monononylphenyl polyoxyethylene phosphate, magnesium saponified monononylphenyl polyoxyethylene phosphate, zinc saponified monononylphenyl polyoxyethylene phosphate, calcium saponified monolauryl polyoxyethylene phosphate, magnesium saponified monolauryl polyoxyethylene phosphate, and zinc saponified monolauryl polyoxyethylene phosphate. These can be used individually or in mixtures of two or more types. The partially saponified organic phosphoric acid esters are also marketed as resin additives and are appropriate for use in terms of cost.

[0052]

The amount in which the fatty acid metal salt (D) or organic phosphoric acid esters (E) is used in the polyester-based resin composition of the present invention is in the range of 0.01-5 parts by weight, preferably 0.02-4 parts by weight, more preferably 0.03-3 parts by weight, per 100 parts by weight of the polyester-based resin (A). The desired effects are not

obtained when the amount in which the fatty acid metal salt (D) or organic phosphoric acid esters (E) is added is less than 0.01 part by weight. The transparency of the sheet or film molded decreases when the admixed amount exceeds 5 parts by weight.

[0053]

Combined use of a fatty acid ester or oxidized polyethylene wax is effective in making it easier for the polyester-based resin composition of the present invention to be released from the metal rolls (calender rolls). However, the fatty acid ester or oxidized polyethylene wax is highly effective in achieving better releasability, so the transparency of the molded sheet or film tends to be compromised and bleedout tends to occur. The amount must therefore be kept to a minimum when this type of additive is introduced. Concretely speaking, 0.5 parts by weight or less per 100 parts by weight of the polyester-based resin (A) is preferred.

[0054]

The number of carbon atoms in the fatty acid moiety of the fatty acid ester used herein is not particularly restricted. However, natural wax or synthetic wax made from an ester of an aliphatic saturated carboxylic acid with 12-28 carbon atoms and an aliphatic saturated alcohol with 2-30 carbon atoms is preferred in consideration of the volatility of the ester obtained. Examples of the aliphatic saturated carboxylic acid with 12-28 carbon atoms in the synthetic wax include lauric acid, myristic acid, stearic acid, behenic acid, lignoceric acid, cerotinic acid, and montanic acid. Examples of the aliphatic saturated alcohol with 2-30 carbon atoms include monohydric alcohols such as ethyl alcohol, octyl alcohol, lauryl alcohol, stearyl alcohol, behenyl alcohol, pentacosyl alcohol, ceryl alcohol, octacosyl alcohol, and myrisyl alcohol; polyhydric alcohols such as dihydric alcohols such as ethylene glycol-1,2-butanediol-1,3-butanediol, 1,4-butanediol, and 2,3-butanediol; and trihydric alcohols such as glycerin.

[0055]

Examples of synthetic waxes include stearyl laurate, stearyl myristate, stearyl stearate, octyl behenate, lauryl behenate, myristyl behenate, stearyl behenate, behenyl behenate, pentacosyl behenate, ceryl lignocerate, octacosyl lignocerate, myrisyl lignocerate, stearyl cerotate, behenyl cerotate, ceryl cerotate, myrisyl cerotate, ethyl montanate, ceryl montanate, and glycol montanate. Examples of natural waxes include montan wax, carnauba wax, beeswax, candelilla wax, rice bran wax, and ibota wax. Glycol montanate, montanic glyceride, and montan

wax among them are preferred from the standpoint of volatility and releasability from the metal rolls. These are marketed as resin additives and are appropriate for use in terms of cost.

[0056]

The type of oxidized polyethylene wax also is not particularly restricted, and either low or high density is acceptable. However, a partially oxidized polyethylene wax with an acid value of 1-40 mg KOH/g and a molecular weight of 10,000 or less in terms of the weight-average molecular weight is preferred in consideration of the releasability of the resin from the metal rolls. These are marketed as resin additives and are appropriate for use in terms of cost.

[0057]

The film or sheet of the present invention is a film or sheet molded by calendering from the aforementioned thermoplastic polyester resin composition for calendering. The film or sheet, when formed by calendering from the thermoplastic polyester resin composition for calendering, has a thickness of 0.2 mm or less and a shrinkage index of less than 10% when heat-treated for 60 minutes at a temperature 10°C higher than the glass transition temperature of the molded article, a shrinkage index of less than 15% when heat-treated for 60 minutes at a temperature 20°C higher than the glass transition temperature of the molded article, and a shrinkage index of less than 20% when heat-treated for 60 minutes at a temperature 40°C higher than the glass transition temperature of the molded article.

[0058]

The glass transition temperature referred to herein is determined as the peak temperature of the complex modulus E'' obtained when measurement is conducted while the temperature is raised from at least 50°C less than the glass transition temperature of the film or sheet using a tensile-type dynamic viscoelastometer. The measurement is carried out at a temperature elevation rate of 2-10°C/min and a frequency in the 5-20 Hz range.

[0059]

A film or sheet with the above properties can be produced from the thermoplastic polyester resin composition for calendering according to the present invention by using calendering equipment in which the temperature of the calender rolls is set from 80°C to less than 150°C, preferably 90°C to less than 140°C, and more preferably 100°C to less than 130°C, higher than the glass transition temperature of the composition of the present invention, and y

taking off the film or sheet onto cooling rolls at a draw ratio of from 130% to less than 500%, preferably 140% to less than 400%, more preferably 150% to less than 350%.

[0060]

The term "draw ratio" used herein refers to the ratio of the surface speed of the cooling rolls to the surface speed of the calender rolls on the molten resin supply side when the molten resin that has been rolled by the calendering machine is taken off by the cooling rolls.

[0061]

Rotation of the mass of molten resin (commonly referred to as "the bank") formed between the calender rolls becomes irregular when the calender roll temperature is less than 80°C above the glass transition temperature of the thermoplastic polyester resin composition of the present invention. It therefore becomes difficult to obtain a transparent film or sheet. The molten resin sticks to the calender rolls and there is a risk that it will become impossible to produce a film or sheet by calendering when the calender roll temperature is more than 150°C higher than the glass transition temperature of the thermoplastic polyester resin composition of the present invention.

[0062]

There is a risk that a film or sheet with a thickness of 0.2 mm or less cannot be produced in a stable manner by calendering when the film or sheet is produced at a draw ratio of less than 130% on the take-off rolls. This is because the gap between the calender rolls must be narrow to produce a film or sheet with a thickness of 0.2 mm or less when the draw ratio is less than 130%. The molten resin composition therefore evolves heat and reaches a high temperature even when the temperature of the calender rolls is set low, and there is a risk that the melt will stick to the calender rolls. Narrowing the gap between the calender rolls also increases the driving power of the calendering equipment and further lowers the draw ratio, resulting in inferior production speed.

On the other hand, problems such as increased shrinkage arise even with the thermoplastic polyester resin composition of the present invention when the draw ratio is more than 500% at the take-off rolls during production of a film or sheet.

[0063]

The thermoplastic polyester resin composition of the present invention can be prepared by simply admixing the following components into the pelleted or powdered polyester-based

resin (A): the organically modified layered silicate (B), the fatty acid with 18 or more carbon atoms (C), and the fatty acid metal salt (D) or organic phosphoric acid esters (E) and, if necessary, the fatty acid ester or oxidized polyethylene wax, or by melt kneading these components in a kneader in advance.

Known equipment can be used herein as the kneader. It is preferable to use equipment such as a single- or twin-screw extruder, kneader, cokneader, planetary mixer, or Banbury mixer for ease of handling and to permit homogeneous dispersion.

[0064]

The polyester-based resin composition of the present invention can also be produced by a method in which a material (commonly referred to as "the master batch") is first prepared by admixing the following components into the polyester-based resin (A) in high concentrations: the organically modified layered silicate (B), the fatty acid (C), the fatty acid metal salt (D) or organic phosphoric acid ester (E), and the fatty acid ester or oxidized polyethylene wax, and then melt kneading the material with the polyester-based resin (A). The kneaders given as examples above can also be used appropriately in the production of the master batch.

[0065]

Conventionally known antioxidants such as hindered phenols, thioethers, amines, and phosphoric acids; conventionally known ultraviolet absorbers such as benzophenones, benzoates, benzotriazoles, cyanoacrylates, and hindered amines; anionic, cationic, and nonionic low-molecular or high-molecular antistatic agents; thickeners such as epoxy compounds and isocyanate compounds; coloring agents such as dyes and pigments; ultraviolet blockers such as titanium oxide and carbon black; reinforcing materials such as glass fibers and carbon fibers; and fillers such as silica, clay, calcium carbonate, barium sulfate, glass beads, and talc, as long as flame retardants, plasticizers, foaming agents, antimicrobials, antifungals, fluorescent whiteners, surfactants, crosslinking agents, and the like can also be added as is appropriate within the range that does not harm the ease of calendering of the polyester-based resin composition of the present invention.

[0066]

Other thermoplastic resins, such as polyolefins, polyvinyl chlorides, polystyrenes, polyethers, polyesters, polyamides, and polyimides, may also be added to the polyester-based

resin composition of the present invention within the range that does not harm the processability in calendering.

[0067]

The polyester-based resin composition of the present invention is suited to the production of a film or sheet by a calendering machine after being melt kneaded in a mixer or a single- or twin-screw extruder. The release of the molten resin from the calender rolls and its take-off by the cooling rolls are good, and the transparency is also good when producing a film or sheet. There is also no bleedout on the calender rolls, and longitudinal streaks caused by air being entrained into the resulting film or sheet are prevented from developing. This polyester-based resin composition therefore makes it possible to mold films and sheets with an excellent appearance at high productivity by calendering.

[0068]

The film or sheet obtained by calendering from the polyester-based resin composition of the present invention has excellent transparency, rigidity, gas barrier properties, and wettability and undergoes little shrinkage. It can therefore be used appropriately as a transparent or colored film or sheet for a cosmetic film or sheet, a film or sheet for cards, a film or sheet for various types of packaging containers, a film or sheet supplied for printing or adhesive processing, a film or sheet processed on the surface by metal deposition or sputtering, a film or sheet used to prevent flying glass or to block heat applied to locations exposed to relatively high temperatures such as windows, a film or sheet for displays, and the like.

[0069]

[Working Examples]

The present invention is explained in greater detail below through concrete working examples. The present invention, however, is not limited to these working examples.

[0070]

The materials used in the working examples and comparative examples will first be explained.

<Polyester-based resin (A)>

A-1: Pelleted polyester-based resin (trade name: Tsunami Copolyester GS2, manufactured by Eastman Chemical Products, Inc.) with a weight-average molecular weight of 25,000 composed of repeating units of a dicarboxylic acid component that is 100 mol%

terephthalic acid and does not contain any other dicarboxylic acid, and a glycol component that is 60-75 mol% ethylene glycol and 40-25 mol% 1,4-cyclohexanedimethanol and does not contain any other glycol component. F-1: Polyethylene terephthalate (trade name: TR8580HP, manufactured by Teijin).

The results obtained by measuring the polyester-based resin A-1 and F-1 at a temperature elevation rate of 20°C/min using a Perkin-Elmer DSC were as shown in Table 1.

[0071]

[Table 1]

Polyester-based resin	Melting peak temperature	Crystal heat of fusion
A-1	Not observed	0 J/g
F-1	245°C	68 J/g

[0072]

<Organically modified layered silicate (B)>

B-1: Organically modified layered silicate prepared according to Reference Example 1

B-2: Organically modified layered silicate prepared according to Reference Example 2

B-3: Organically modified layered silicate prepared according to Reference Example 3

B-4: Organically modified layered silicate (trade name: Cloisite 10A, manufactured by Southern Clay Products) with 39% by weight dimethylbenzyl hydrogenated tallow ammonium ions introduced by ion exchange between the layers of montmorillonite with a cation exchange capacity of 125 mEq/100 g.

B-5: Organically modified layered silicate (trade name: Cloisite 15A, manufactured by Southern Clay Products) with 43% by weight dimethyl dihydrogenated tallow ammonium ions introduced by ion exchange between the layers of montmorillonite with a cation exchange capacity of 125 mEq/100 g.

B-6: Organically modified layered silicate (trade name: Cloisite 30B, manufactured by Southern Clay Products) with 30% by weight methyl di-tallow alkyl bis-2-hydroxyethyl ammonium ions introduced by ion exchange between the layers of montmorillonite with a cation exchange capacity of 90 mEq/100 g.

<Layered silicate>

G-1: High-purity montmorillonite (trade name: Ben-gel A, manufactured by Hojun)

[0073]

[Reference Example 1]

3 kg of natural Na montmorillonite (trade name: Kunipia F, manufactured by Kunimine Kogyo; cation exchange capacity: 120 mEq/100 g) were dispersed by stirring in 100 L of warm water. After 1.0 times the cation exchange capacity of dimethyl stearyl benzyl ammonium salt was added thereto and the system was stirred for 1 hour, the precipitate produced was filtered out and washed with warm water. Washing and filtration were repeated three times. The solid obtained was then dried to obtain organically modified layered silicate B-1.

[0074]

[Reference Example 2]

3 kg of natural Na montmorillonite (trade name: Kunipia F, manufactured by Kunimine Kogyo; cation exchange capacity: 120 mEq/100 g) were dispersed by stirring in 100 L of warm water. After 1.0 times the cation exchange capacity of benzalkonium salt was added thereto and the system was stirred for 1 hour, the precipitate produced was filtered out and washed with warm water. Washing and filtration were repeated three times. The solid obtained was then dried to obtain organically modified layered silicate B-2.

[0075]

[Reference Example 3]

3 kg of natural Na montmorillonite (trade name: Ben-gel A, manufactured by Hojun; cation exchange capacity 94 mEq/100 g) were dispersed by stirring in 100 L of warm water. After 1.0 times the cation exchange capacity of dimethyl distearyl ammonium salt was added thereto and the system was stirred for 1 hour, the precipitate produced was filtered out and washed with warm water. Washing and filtration were repeated three times. The solid obtained was then dried to obtain organically modified layered silicate B-3.

The percent by weight of ash (ash(%)), as determined from the weight of the ash that remained after 10 g each of the organically modified layered silicates B-1 to B-5 and layered silicate G-1 was weighed out and heated for 30 minutes in a 1000°C electric furnace, is shown in Table 2.

[0076]

[Table 2]

Organically modified layered silicate or layered silicate	% by weight ash (ash (%))
B-1	60% by weight
B-2	65% by weight
B-3	62% by weight
B-4	61% by weight
B-5	57% by weight
B-6	70% by weight
G-1	92% by weight

[0077]

<Fatty acid with 18 or more carbon atoms (C)>

C-1: Montanic acid (trade name: Licowax S, manufactured by Clariant Japan)

<Fatty acid metal salt (D)>

D-1: Calcium montanate (trade name: Licomont CaV102, manufactured by Clariant Japan)

<Organic phosphoric acid esters (E)>

E-1: Mixture of monostearyl phosphate and distearyl phosphate in a 6:4 weight ratio (trade name: AX-518, manufactured by Daikyo Kasei Kogyo)

[0078]

<Other additives>

Fatty acid ester: Ester of montanic acid and 1,4-butanediol (trade name: Licowax E, manufactured by Clariant Japan)

Oxidized polyethylene wax: Wax with an acid value of 15-19 mg KOH/g (trade name: Licowax PED191, manufactured by Clariant Japan)

Antioxidant: Tetrakis[methylene-3-(3',5'-di-t-butyl-4-hydroxyphenyl)propionate]methane (trade name: Adekastab, manufactured by Asahi Denka)

[0079]

The values listed in Tables 3 and 4 will be explained next.

The "amount of silicate per 100 parts by weight of resin" and "compounding agent C-1/silicate" were determined as follows.

"Amount of silicate (b') per 100 parts by weight of resin"

The "amount of silicate (b') per 100 parts by weight of polyester-based resin (A)" was determined in terms of parts by weight on the basis of the parts by weight (b'') of organically modified layered silicate (B) or layered silicate added per 100 parts by weight of polyester-based resin (A), and on the basis of the percent by weight of ash (ash (%)) of the organically modified layered silicate (B) or layered silicate shown in Table 2 using the following formula, because the organically modified layered silicate (B) and layered silicate contained components other than silicate, such as water and organic onium ions introduced by ion exchange.

$$b' = b'' \times \text{ash} (\%) / 100$$

"Compounding agent C-1/silicate"

The "compounding agent C-1/silicate" is the "amount (c') of compounding agent C-1 added per 100 parts by weight of resin," as determined using the formula described above (in parts by weight) divided by the "amount of silicate (b') per 100 parts by weight of resin " (in parts by weight). The ratio was determined as follows.

Compounding agent C-1/silicate (parts by weight/parts by weight) = c'/b'

The other values and methods of evaluation are explained in the working and comparative examples.

[0080]

<Working Examples 1-8>

The compounding agents were mixed in the parts by weight shown in Table 3, and compositions were obtained by melt kneading the components for 10 minutes at 180°C using a Laboplastomill mixer (R-60) manufactured by Toyo Seiki. The glass transition temperatures of sheets obtained by press molding these compositions in a thickness of 1 mm at 180°C were determined as the peak temperature of the complex modulus E'' measured at a temperature elevation rate of 2°C/min and a frequency of 10 Hz using a Rheograph manufactured by Toyo Seiki. The result in all of the sheets was 80°C.

Next, the compounding agents were mixed in the parts by weight shown in Table 3. After the components had been mixed until homogeneous in a Henschel mixer, polyester-based resin compositions were prepared by kneading in a Banbury mixer until the resin temperature reached the 160-175°C range. These were rolled using a reverse L-shaped four-roll calendering machine set at 190°C, which is 110°C higher than the glass transition temperature of the composition,

taken off at a draw ratio of 150% or 250[%], and cooled to produce sheets that were 1000 mm wide and 100 μ m thick.

The glass transition temperatures of the sheets obtained by calendering were determined as the peak temperature of the complex modulus E'' measured at a temperature elevation rate of 2°C/min and a frequency of 10 Hz using a Rheograph manufactured by Toyo Seiki. The result in all sheets was 80°C, as in the compositions.

[0081]

The evaluations shown in Table 3 were performed by the following methods.

[Evaluation of dispersiveness of the organically modified layered silicate, and evaluation of the releasability from metal]

The melt was examined immediately after a melt kneading operation that lasted for 10 minutes and was performed at 180°C using a Laboplastomill mixer (R-60) manufactured by Toyo Seiki prior to molding by calendering, and the state of dispersion of the organically modified layered silicate (B) in the molten state was evaluated. The state of dispersion of the organically modified layered silicate (B) in the molded article was also evaluated by the visual inspection of the molded article when 1-mm thick molded articles were produced by subjecting the melt kneaded compound to compression molding at 180°C. The state of dispersion of the organically modified layered silicate (B) was taken to be "○" when grains of the organically modified layered silicate (B) could not be observed in the melt or the molded article, and as "×" when the grains were observed.

Releasability from metal was evaluated by assessing the ease with which the melt from the mixer could be released during a pre-kneading operation that lasted for 10 minutes and was performed at 180°C using a Laboplastomill mixer (R-60) manufactured by Toyo Seiki. The releasability was taken to be "○" when the melt could be released easily from the mixer after kneading, and as "×" when the melt adhered and could not be released from the mixer.

[0082]

[Evaluation of roll processability, surface state of the sheet, and flexibility of the sheet]

A compound obtained by pre-kneading that lasted for 10 minutes and was performed at 180°C using a Laboplastomill mixer (R-60) manufactured by Toyo Seiki before molding in a calendering machine was kneaded using two 10-inch rolls heated to 190°C to obtain a sheet

thickness of 150 μm , and the adhesiveness of the compound to the rolls and the state of the bank were evaluated.

The adhesiveness to the rolls was taken to be "○" when the melt did not adhere to the roll surface and could be processed, and as "×" when the melt stuck to the roll surface. The state of the bank was taken to be "○" when the bank was spindle-shaped, no air was entrained into the bank, the bank surface was smooth, and the bank rotated smoothly, and as "×" when the bank was not spindle-shaped, air was entrained into the bank, the surface was nappy, or the bank did not rotate smoothly.

The sheet take-off property was taken to be "○" when the sheet was removed from the roll during kneading and could be easily taken off, and as "×" when the sheet stretched and could not be taken off.

To evaluate the surface state of the sheet, samples were prepared by laminating the sheet with release paper, and the surface state of the sheet was examined visually. The surface state of the sheet was taken to be "○" when there were no longitudinal streaks caused by incorporation of air, the surface was smooth, and a transparent sheet was obtained, and as "×" when there were longitudinal streaks caused by incorporation of air or when the sheet was opaque.

To evaluate the flexibility of the sheet, a 10-cm² sheet was cut from the samples prepared by lamination with release paper. The flexibility was taken to be "○" when the sheet did not crack when bent at a 90° angle, and as "×" when the sheet cracked.

[0083]

[Measurement of melt tension]

The melt tension was measured to evaluate the ease of take-off from the cooling roll during calendering before molding in a calendering machine. The compound obtained by melt kneading for 10 minutes at 180°C using a Laboplastomill mixer (R-60) manufactured by Toyo Seiki was measured at a temperature of 190°C, a cross head speed of 5 mm/min, and a winding rate of 20 m/min using a Capirograph manufactured by Toyo Seiki and equipped with an L/D = 20/1 mm capillary.

[0084]

[Measurement of melt viscosity]

The melt viscosity was measured to evaluate the magnitude of electrical power required to drive the calendering equipment during calendering before molding in a calendering machine.

The compound obtained by melt kneading for 10 minutes at 180°C using a Laboplastomill mixer (R-60) manufactured by Toyo Seiki was measured at a temperature of 190°C and a shear rate of 250/sec using a Capirograph manufactured by Toyo Seiki and equipped with an L/D = 20/1 mm capillary.

[0085]

[Measurement of shrinkage]

A 15-cm² sheet was cut from the sheet obtained by calendering, and the lengthwise shrinkage of the sheet was determined from the change in the length of the sheet before and after a heat treatment that lasted for 60 minutes in an oven set at a prescribed temperature (listed in Tables 3 and 4), followed by the removal of the sheet.

[0086]

[Tensile test]

Test pieces the size of small cards 200 mm long and 19 mm wide were punched out from the sheets obtained by calendering so that the lengthwise direction would form the long side. A tensile test was conducted according to JIS K-7127.

[0087]

[Measurement of oxygen permeability coefficient]

Measurement was conducted with the aid of a differential pressure-type gas permeability tester manufactured by Toyo Seiki using sheets obtained by calendering.

[0088]

[Measurement of wetting tension]

The wetting tension of the sheet surface by a wetting reagent was measured using the sheets obtained by calendering.

Table 3 summarizes the results of evaluation.

[0089]

<Comparative Example 1>

A composition was obtained by melting and kneading components for 10 minutes at 180°C in a Laboplastomill mixer (R-60) manufactured by Toyo Seiki. The glass transition temperature of the sheet obtained by press molding this composition to a thickness of 1 mm at 180°C was determined as the peak temperature of the complex modulus E" measured at a

temperature elevation rate of 2°C/min and a frequency of 10 Hz using a Rheograph manufactured by Toyo Seiki. The result was 80°C.

Next, the compounding agents were added in the parts by weight shown in Table 4. After the components had been mixed until homogeneous in a Henschel mixer, a polyester resin composition was prepared by kneading in a Banbury mixer until the resin temperature reached the 160-175°C range. The composition was rolled using a reverse L-shaped four-roll calendering machine set at 190°C, which is 110°C higher than the glass transition temperature of the composition, and taken off at a draw ratio of 150% or 250%. However, the sheet stretched and could not be taken off by the cooling rolls. It was therefore taken off at a draw ratio of 350%, and a 1000-mm wide, 100-μm thick sheet was produced by cooling. The electrical power required to drive the molding machine during calendering of this composition was 50% greater than that in Working Examples 1, 2, and 5-7; 20% greater than in Working Example 3; 70% greater than in Working Example 4; and 30% greater than in Working Example 8.

The glass transition temperature of the resulting sheet was determined as the peak temperature of the complex modulus E'' measured at a temperature elevation rate of 2°C/min and a frequency of 10 Hz using a Rheograph manufactured by Toyo Seiki. It was 80°C, the same as the composition.

The results of evaluations performed before molding in a calendering machine are shown in Table 4 together with the results of evaluation of the sheet produced by calendering machine.

The evaluations shown in Table 4 were conducted using the same methods and criteria as in the working examples.

It is understood from this comparative example that the melt tension is lower than in the working examples and that the take-off properties are inferior when roll processability is evaluated for a sheet to which the organically modified layered silicate (B) is not added. Take-off was not possible without raising the draw ratio during calendering as well, and the take-off properties of the sheet were inferior to that of the working examples.

The melt viscosity was also higher than in the working examples, and the electrical power required to drive the molding machine during calendering was higher than in the working examples. Furthermore, the shrinkage, tensile properties, oxygen barrier property, and wettability were understood to be inferior to those of the sheets shown in the working examples in Table 3.

[0090]

<Comparative Example 2>

The dispersiveness of the organically modified layered silicate (B), releasability from metal, melt tension, melt viscosity, roll processability, surface state of the sheet, and flexibility of the sheet were evaluated by the same methods as in the working examples using a composition in which the compounding agents were added in the parts by weight shown in Table 4.

A large amount of air was entrained into the bank in the composition of Comparative Example 2 that did not contain a fatty acid metal salt or a phosphoric acid ester. The air was transferred to the sheet as longitudinal streaks, and the surface state of the sheet was inferior. The thickness was also uneven. The other evaluations, including calendering, therefore could not be performed.

The evaluation results are shown in Table 4. The evaluations shown in Table 4 were conducted using the same methods and criteria as in the working examples.

It is understood from this comparative example that a sheet with a superior surface state and evenness of thickness is not obtained because air is incorporated into the bank when a fatty acid metal salt or phosphoric acid ester is not added.

[0091]

<Comparative Example 3>

The dispersiveness of the organically modified layered silicate (B), releasability from metal, roll processability, surface state of the sheet, and flexibility of the sheet were evaluated by the same methods as in the working example using a composition in which the compounding agents were added in the parts by weight shown in Table 4.

The sheet cracked during evaluation of the flexibility of the sheet in the case of the composition of Comparative Example 3 in which the organically modified layered silicate (B) was added in a quantity exceeding 20 parts by weight per 100 parts by weight of the polyester-based resin (A). The other evaluations, including calendering, therefore could not be performed.

The evaluation results are shown in Table 4. The evaluations shown in Table 4 were conducted using the same methods and criteria as in the working examples.

It is understood from this comparative example that the sheet becomes brittle when the organically modified layered silicate (B) is added in a quantity exceeding 20 parts by weight per 100 parts by weight of the polyester-based resin (A).

[0092]

<Comparative Example 4>

The dispersiveness of the organically modified layered silicate (B), releasability from metal, and roll processability were evaluated by the same methods as in the working examples using a composition in which the compounding agents were added in the parts by weight shown in Table 4.

The state of dispersion of the organically modified layered silicate in the molded article could not be evaluated because the composition of Comparative Example 3 [sic] in which the fatty acid with 18 or more carbon atoms (C) was added in a quantity exceeding 10 parts by weight per 100 parts by weight of the polyester-based resin (A) turned white when cooled. In evaluation of the roll processability, the molten resin slipped on the roll surface. A sheet could not be formed and the bank also was not spindle-shaped. The surface state of the sheet and the flexibility of the sheet consequently could not be evaluated. The other evaluations, including calendering, also could not be performed.

The evaluation results are shown in Table 4. The evaluations shown in Table 4 were conducted using the same methods and criteria as in the working examples.

It is understood from this comparative example that not only is a sheet with inferior transparency obtained, but also roll processing itself becomes impossible due to excessive slippage when the fatty acid with 18 or more carbon atoms (C) is added in a quantity exceeding 10 parts by weight per 100 parts by weight of the polyester-based resin (A).

[0093]

<Comparative Examples 5 and 6>

The dispersiveness of the organically modified layered silicate (B) and the releasability from metal were evaluated by the same methods as in the working examples using compositions in which the compounding agents were added in the parts by weight shown in Table 4.

The melt of these compositions could not be released from the metal and turned white when cooled. The state of dispersion of the organically modified layered silicate in the molded article consequently could not be evaluated, and the other evaluations, including calendering, could not be performed.

The evaluation results are shown in Table 4. The evaluations shown in Table 4 were conducted using the same methods and criteria as in the working examples.

It is understood from the results of these valuations that not only is a sheet of inferior transparency obtained, but the melt also cannot be released from metal when the fatty acid metal salt (D) or organic phosphoric acid ester (E) is added in a quantity that exceeds 5 parts by weight per 100 parts by weight of the polyester-based resin (A).

[0094]

<Comparative Examples 7-10>

The dispersiveness of the organically modified layered silicate and releasability from metal were evaluated by the same methods as in the working examples using compositions in which the compounding agents were added in the parts by weight shown in Table 4.

As a result, numerous grains of organically modified layered silicate were seen in the melt, and the sheets also became opaque during press molding. The other evaluations, including calendering, consequently were not performed.

The evaluation results are shown in Table 4. The evaluations shown in Table 4 were conducted using the same methods and criteria as in the working examples.

It is understood from the results of these evaluations that the dispersion of the organically modified layered silicate (B) becomes inadequate and only a sheet with inferior transparency is obtained when no fatty acid with 18 or more carbon atoms (C) is added, as in Comparative Examples 7 and 8, and also when the ratio (c'/b') of the parts by weight (b') of silicate in the organically modified layered silicate added per 100 parts by weight of the polyester-based resin (A) and the parts by weight (c') of fatty acid with 18 or more carbon atoms added per 100 parts by weight of the polyester-based resin (A) is less than 0.1, as in Comparative Example 9. The dispersion of the layered silicate is also inadequate and only a sheet with inferior transparency is obtained when a layered silicate that has not been organically modified is used, as in Comparative Example 10.

[0095]

<Comparative Examples 11 and 12>

The compounding agents were mixed in the parts by weight shown in Table 4 and melt kneaded for 10 minutes at 180°C using a Laboplastomill mixer (R-60) manufactured by Toyo Seiki. However, the polyester-based resin (F-1) did not melt. Numerous grains of an organically modified layered silicate were seen in the melt when the state of dispersion of the organically modified layered silicate was evaluated in a molten state by examining the melt immediately

after kneading had been conducted for 10 minutes by setting the kneading temperature to 270°C. The molten composition also could not be released from metal.

The state of dispersion of the organically modified layered silicate in the molded article could not be evaluated because the molten composition that contained the polyester-based resin F-1 turned white when cooled. The other evaluations, including calendering, also could not be performed.

The evaluation results are shown in Table 4. The evaluations shown in Table 4 were conducted using the same methods and criteria as in the working examples.

It is understood from this comparative example that the dispersion of the organically modified layered silicate becomes inadequate and the melt also cannot be released from metal when the crystalline polyester-based resin used has a crystal melting peak that exceeds 200°C and a crystal heat of fusion that exceeds 50 J/g, as measured by DSC.

[0096]

[Table 3]

			(Unit)	WE 1	WE 2	WE 3	WE 4	WE 5	WE 6	WE 7	WE 8
Compounding agent	Polyester-based resin (A)	A-1 (parts by weight)	100	100	100	100	100	100	100	100	100
	Polyester-based resin	F-1 (parts by weight)									
	Organically modified layered silicate (B)	B-1 (parts by weight)	8.3								
		B-2 (parts by weight)						7.7			
		B-3 (parts by weight)							8.1		
		B-4 (parts by weight)								8.2	
		B-5 (parts by weight)				13.2					5.2
		B-5 (parts by weight)		7.1	2.1						
	Layered silicate	G-1 (parts by weight)									
	Fatty acid (C)	C-1 (parts by weight)	1.5	1.5	0.5	5.0	1.25	1.5	1.0	2.0	
	Fatty acid metal salt (D)	D-1 (parts by weight)	0.07	0.1		1.5	0.1		0.5	0.1	
	Organic phosphoric acid esters (E)	E-1 (parts by weight)			0.1			0.5			
	Other additives	Fatty acid ester (parts by weight)			0.05						
		Oxidized polyethylene wax (parts by weight)							0.05		
Antioxidant (parts by weight)		0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	
Amount (b') of silicate per 100 parts by weight of resin (parts by weight)			5.0	5.0	1.5	7.5	5.0	5.0	5.0	3.0	
Compounding agent C-1/silicate (c'/b') (parts by weight/parts by weight)			0.30	0.30	0.33	0.67	0.25	0.30	0.20	0.67	
Results of evaluation	State of dispersion of layered silicate	Melt	○	○	○	○	○	○	○	○	○
		Molded article	○	○	○	○	○	○	○	○	○
	Release from metal			○	○	○	○	○	○	○	○
	Roll processability	Stickiness	○	○	○	○	○	○	○	○	○
		State of bank	○	○	○	○	○	○	○	○	○
		Sheet take-off	○	○	○	○	○	○	○	○	○
	Surface state of sheet		○	○	○	○	○	○	○	○	○
	Flexibility of sheet		○	○	○	○	○	○	○	○	○
	Melt tension (g)		2.6	2.6	2.0	2.7	2.5	2.6	2.5	2.5	2.3
	Melt viscosity (Pa·s)		1000	1000	1400	800	1000	1000	1000	1000	1200
Draw ratio during calendering (%)			150	250	250	250	150	250	150	250	

		(Unit)	WE 1	WE 2	WE 3	WE 4	WE 5	WE 6	WE 7	WE 8
Results of evaluation	Shrinkage (%) (heat treatment for 60 min)	90°C	2.0	5.0	8.0	3.0	2.0	5.5	2.0	7.0
		100°C	3.5	6.5	13.0	4.5	3.5	6.5	4.0	10.5
		120°C	5.0	10.5	18.0	7.0	5.5	11.0	5.5	15.0
	Tensile test	Modulus (MPa)	1600	1700	1300	2000	1600	1700	1600	1500
		Yield stress (MPa)	65.4	66.7	55.2	70.2	65.6	67.1	65.9	59.5
	Oxygen permeability coefficient (mL·mm/(m ² ·24 hr·atm))		2.9	3.0	4.5	2.6	2.9	3.0	3.1	3.5
	Wet tension (mN/m)		39	39	37	40	38	39	39	38

[0097]
[Table 4]

			(Unit)	CE 1	CE 2	CE 3	CE 4	CE 5	CE 6	CE 7	CE 8	CE 9	CE 10	CE 11	CE 12	
Compounding agent	Polyester-based resin (A)	A-1 (parts by weight)	100	100						100	100	100	100			
	Polyester-based resin	F-1 (parts by weight)			100	100	100	100						100	100	
	Organically modified layered silicate (B)	B-1 (parts by weight)		8.3	30.0	8.3	8.3	8.3	8.3	8.3	4.2			8.3	8.3	
		B-2 (parts by weight)														
		B-3 (parts by weight)														
		B-4 (parts by weight)														
		B-5 (parts by weight)														
	Layered silicate	G-1 (parts by weight)											5.4			
	Fatty acid (C)	C-1 (parts by weight)	1.0	1.5	5.0	20.0	1.5	1.5				0.09	1.5	1.5	1.5	
	Fatty acid metal salt (D)	D-1 (parts by weight)	0.1		0.1	0.1		10.0	0.1	2.0	0.009	0.1	0.1			
	Organic phosphoric acid esters (E)	E-1 (parts by weight)					10.0								0.1	
	Other additives	Fatty acid ester (parts by weight)														
		Oxidized polyethylene wax (parts by weight)														
		Antioxidant (parts by weight)	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Amount (b') of silicate per 100 parts by weight of resin (parts by weight)			0	5.0	18.0	5.0	5.0	5.0	5.0	5.0	2.5	5.0	5.0	5.0		
Compounding agent C-1/silicate (c'/b') (parts by weight/parts by weight)			—	0.30	0.28	4.00	0.30	0.30	0	0	0.036	0.3	0.3	0.3		

(Unit)			CE 1	CE 2	CE 3	CE 4	CE 5	CE 6	CE 7	CE 8	CE 9	CE 10	CE 11	CE 12
Results of evaluation	State of dispersion of layered silicate	Melt	—	○	○	○	○	○	×	×	×	×	×	×
		Molded article	—	○	○				×	×	×	×		
	Release from metal		○	○	○	○	×	×	×	○	×	○	×	×
	Roll processability	Stickiness	○	○	○	○								
		State of bank	○	×	○	×								
		Sheet take-off	×	○	○									
	Surface state of sheet		○	×	○									
	Flexibility of sheet		○	○	×									
	Melt tension (g)		1.5	2.5										
	Melt viscosity (Pa·s)		1800	1000										
Results of evaluation	Draw ratio during calendering (%)		350											
	Shrinkage (%) (heat treatment for 60 min)	90°C	16.0											
		100°C	19.0											
		120°C	26.0											
	Tensile test	Modulus (MPa)	1000											
		Yield stress (MPa)	49.1											
	Oxygen permeability coefficient (mL·mm/(m ² ·24 hr·atm))		6.6											
	Wet tension (mN/m)		35											

[0098]

[Effect of the Invention]

As is evident from the results of the evaluations shown in Tables 3 and 4, the polyester-based resin composition for calendering of the present invention is well suited to calendering in terms of releasability from metal, rotation properties of the bank, take-off properties of the sheet, and the ability to reduce the electrical power required to drive the equipment during calendering. The composition yields a transparent molded article, and the surface condition of the sheet or film obtained by calendering is good. There is also little shrinkage, and the gas barrier properties and wettability are excellent.

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